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# Evidence of Enhanced Hypersensitive Transition in Erbium-Doped Fibers With Different Al<sub>2</sub>O<sub>3</sub> Content

Shunsuke Ono and Setsuhisa Tanabe

**Abstract**—Precise spectroscopic absorption measurements of erbium-doped aluminosilicate fibers with different Al<sub>2</sub>O<sub>3</sub> content were performed with the Judd–Ofelt analysis. From the Judd–Ofelt analysis, the  $\Omega_2$  parameters of Er<sup>3+</sup> ions in these fibers were found to be about three times as large as those in aluminosilicate bulk glasses. The enhancement of the  $\Omega_2$  parameters led to much stronger line strength of hypersensitive transitions in a fiber form than in a bulk glass form. This indicates that the distortion of the ligand field around the Er<sup>3+</sup> ions are more enhanced in a fiber form than in a bulk glass form. Furthermore, the  $\Omega_6$  and the  $\Omega_2$  parameters increased with an increase of  $q$  content up to 20 000 ppm. This Al<sub>2</sub>O<sub>3</sub>-content dependence of the  $\Omega_6$  parameter was consistent with that of the line strength and the spontaneous emission probabilities of the transition corresponding to the  $^4I_{13/2} \rightarrow ^4I_{15/2}$ .

**Index Terms**—Erbium, glass, optical amplifiers, optical materials, optical spectroscopy.

## I. INTRODUCTION

SILICA-BASED erbium-doped fiber amplifier (EDFA) has been a key device in the telecommunication system based on wavelength division multiplexing (WDM). This is because EDFA uses the emission transition corresponding to the  $^4I_{13/2} \rightarrow ^4I_{15/2}$  of Er<sup>3+</sup> ion and can amplify the light signal in the C-band wavelength region (1530–1570 nm), which is consistent with the low-loss wavelength region of telecom silica fibers.

On the other hand, there is urgent demand for optical amplifiers with a wide and flat gain spectrum due to the rapid increase of information traffic. Many studies about nonsilicate based optical amplifier for new bands (S-band: 1450–1530 nm, L-band: 1570–1630 nm) have been performed in order to expand transmission bandwidth [1], [2]. However, the silica-based erbium-doped fiber (EDF) is still a promising candidate, not only for the C-band amplifier, but also for the S-band and L-band ones because of its high reliability and favorable properties for the optical transmission system. Today, many efforts to modify the properties of silica-based EDF are being made for application to new band amplifiers [3], extracting more potential abilities of erbium-doped silica-based glasses for advanced EDF. So far, spectroscopy of erbium-doped bulk glasses has been well studied in order to improve the properties of EDF [4]. However, as far as we know, few spectroscopic studies have been carried out based on the Judd–Ofelt analysis for Er<sup>3+</sup> ions in fiber forms.

Therefore, this study focuses on the precise spectroscopy and the Judd–Ofelt analysis of aluminosilicate EDF with different Al<sub>2</sub>O<sub>3</sub> content in order to investigate the ligand field structure around the Er<sup>3+</sup> ions in fiber forms. The basic optical properties of EDF with different Al<sub>2</sub>O<sub>3</sub> content were investigated mainly by optical absorption measurements and analyzed precisely by the Judd–Ofelt theory. The effect of the Al<sub>2</sub>O<sub>3</sub> content on the change of the ligand field in aluminosilicate EDFs will be discussed based on the coordination number of Al<sup>3+</sup> in aluminosilicate EDF.

## II. EXPERIMENTAL

### A. Absorption Measurements of EDF

Four EDF samples with different Al<sub>2</sub>O<sub>3</sub> content were used for investigating the compositional dependence of optical properties of EDF. The concentrations of Er<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub> of four EDF samples are shown in Table I. The solid-state and optical parameters for Al<sub>2</sub>O<sub>3</sub>-doped silica-based EDF samples were used in the calculations for the cross section analysis and Judd–Ofelt analysis. The absorption spectrum of an aluminosilicate bulk glass whose composition was the same as that of an aluminosilicate EDF sample (Al<sub>2</sub>O<sub>3</sub> content: 20 000 ppm) was also examined. The refractive index of EDF samples at 1310 nm was measured using a reflectometer, (Ando, AQ7413). Absorption spectra of EDF in the 1.5- $\mu$ m region were measured with three tunable laser sources (Santec, TSL210) in the range of 1420–1640 nm and an optical spectrum analyzer (OSA) (Anritsu, MS9780A). The signal input power was  $-30$  dBm in order not to saturate fiber loss. In the 1.5- $\mu$ m region absorption measurement, fiber lengths of four EDFs were 1 m. The TSL and OSA were controlled by a personal computer using general purpose interface bus (GPIB). Loss was determined by detecting the peak power of a signal output power and subtracting a signal output power from a signal input power.

In order to carry out the Judd–Ofelt analysis, absorption measurements in the wavelength region from 400 to 1650 nm were also performed using a white light source (Ando, AQ4303B), a fiber-optic spectrometer (Ocean optics, USB2000), and the OSA.

### B. Emission Measurements and Cross Section Analysis Based on McCumber Theory

Spontaneous emission of an EDF sample (Al<sub>2</sub>O<sub>3</sub>:20 000 ppm) was measured as side fluorescence at room temperature with a monochromator (Nikon G250) and a photodetector (Electro Optical Systems). Each EDF was pumped by a 980-nm laser diode (LD). The pump power was 150 mW, which was

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TABLE I  
BASIC SOLID-STATE PARAMETERS FOR EDF SAMPLES

sample	$\text{Er}_2\text{O}_3$ (ppm)	$\text{Al}_2\text{O}_3$ (ppm)	$\rho_{\text{Er}_2\text{O}_3}$ ( $\times 10^{18} \text{cm}^{-3}$ )	$n_{1.3 \mu\text{m}}$ (-)	$\lambda_{\text{mean}}(\text{nm})$ ( $1.5 \mu\text{m}$ region)
EDF1	780	5000	5.21	1.476	1523.8
EDF2	880	10000	5.53	1.475	1525.3
EDF3	630	20000	4.10	1.474	1524.6
EDF4	800	40000	5.44	1.481	1523.9

enough power to achieve the population inversion. An experimental emission cross section was calculated using the obtained emission spectrum. In addition, a theoretical emission cross section of the EDF sample ( $\text{Al}_2\text{O}_3$ :20 000 ppm) was also calculated using the measured absorption cross section  $\sigma_a$  through the McCumber relation [5] as follows:

$$\sigma_e(\nu) = \sigma_a \exp \left[ \frac{(E_0 - h\nu)}{kT} \right] \cdot \frac{1 + \sum_{j=2}^8 7 \exp \left( -\frac{\Delta E_{1j}}{kT} \right)}{1 + \sum_{j=2}^7 6 \exp \left( -\frac{\Delta E_{2j}}{kT} \right)} \quad (1)$$

where  $E_0$  is the separation energy between the lowest component of each manifold and assumed to be  $6535 \text{ cm}^{-1}$  ( $\lambda = 1530 \text{ nm}$ ).  $\Delta E_{1j}$  and  $\Delta E_{2j}$  are the differences in energy between the  $j$ th and the lowest component of the  $^4\text{I}_{13/2}$  and the  $^4\text{I}_{15/2}$  level. Here, we supposed that the adjacent components in the same manifold are separated by the same energy  $\delta E_1$  and  $\delta E_2$ . Comparison of the experimental cross section with the theoretical one was performed in the next section.

### III. RESULTS AND DISCUSSION

#### A. $\text{Al}_2\text{O}_3$ Dependence of Absorption Cross Section

Fig. 1(a) shows the absorption cross section  $\sigma_a$  calculated from the loss spectra.  $\sigma_a$  showed the  $\text{Al}_2\text{O}_3$  dependence and increased about double with the increasing content of  $\text{Al}_2\text{O}_3$  up to 20 000 ppm. Fig. 1(b) shows the comparison of the experimental emission cross section for  $\text{Al}_2\text{O}_3$ -doped EDF ( $\text{Al}_2\text{O}_3$ :20 000 ppm) with theoretical one. The theoretical emission cross section was calculated from the absorption cross section using McCumber relation mentioned above. The shape of the theoretical cross section shows good agreement with that of experimental one.

Fig. 2 shows the line strength  $S_{JJ'}$  corresponding to the  $1.5\text{-}\mu\text{m}$  transition and the spontaneous probability  $A_{JJ'}$  of the  $^4\text{I}_{13/2} - ^4\text{I}_{15/2}$  transition and the measured lifetime of the  $^4\text{I}_{13/2}$ ,  $\tau_f$ .

According to the Judd–Ofelt theory, the line strength  $S_{JJ'}$  is related to the integrated absorption cross section of the  $1.5\text{-}\mu\text{m}$  transition in Fig. 1(b) as follows:

$$S_{JJ'} = \frac{3hc(2J+1)}{8\pi^3 e^2 \lambda_{\text{mean}} \rho} n \left[ \frac{9}{(n^2+2)^2} \right] \int_{\text{band}} k(\lambda) d\lambda \quad (2)$$

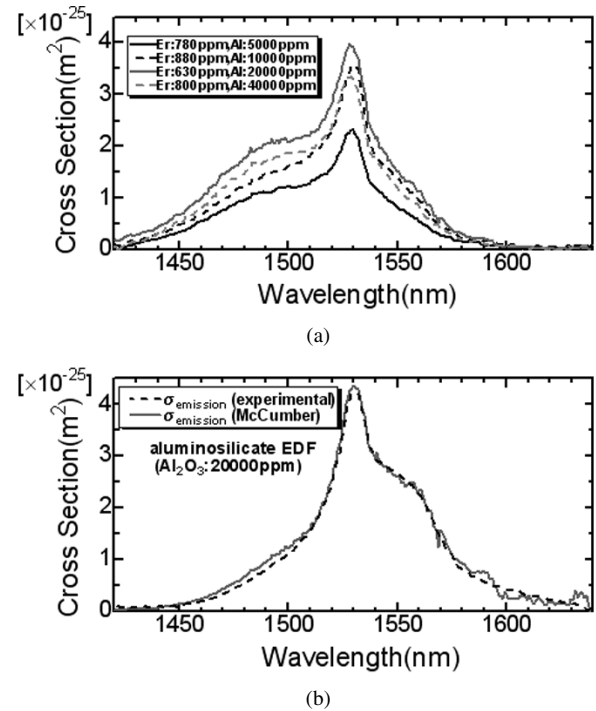


Fig. 1.(a) Absorption cross-section spectra for  $\text{Al}_2\text{O}_3$ -doped EDF in a  $1.5\text{-}\mu\text{m}$  region. (b) Comparison of the experimental emission spectra for  $\text{Al}_2\text{O}_3$ -doped EDF ( $\text{Al}_2\text{O}_3$ :20 000 ppm) with the theoretical one based on McCumber theory.

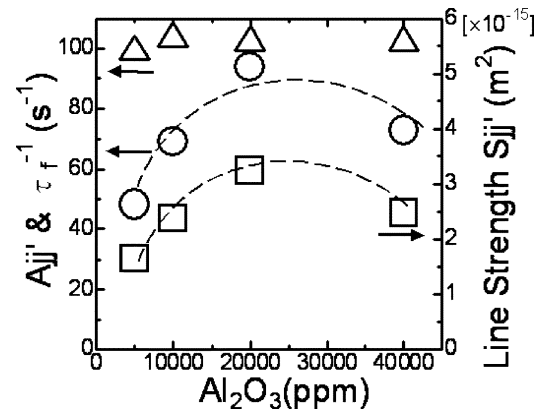


Fig. 2.  $\text{Al}_2\text{O}_3$  dependence of  $S_{JJ'}$  (open square),  $A_{JJ'}$  (open circle), and  $\tau_f$  (open triangle).

where  $k(\lambda)$  is the absorption coefficient at  $\lambda$ ,  $\lambda_{\text{mean}}$  is the mean wavelength of the absorption band,  $\rho$  is the concentration of  $\text{Er}^{3+}$  ions per unit volume,  $h$  is Planck's constant,  $c$  is the

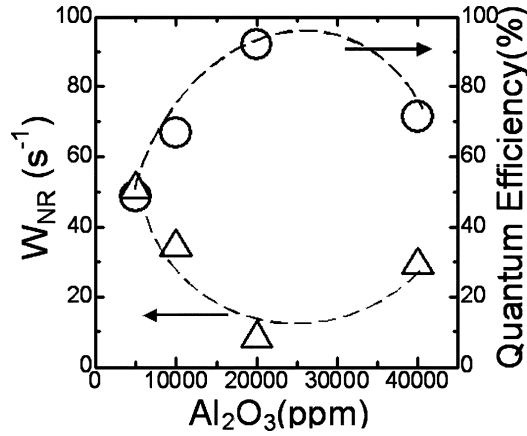


Fig. 3.  $\text{Al}_2\text{O}_3$  dependence of  $W_{\text{NR}}$  and QE.

speed of light,  $e$  is the elementary charge, and  $n$  is the refractive index. The refractive indexes of the core of the EDF were about 1.47 and showed little  $\text{Al}_2\text{O}_3$ -content dependence as shown in Table I.  $J$  is the total angular momentum of the initial state, which is the  $^4\text{I}_{15/2}$ , and  $J'$  is that of the final state  $^4\text{I}_{13/2}$  for the  $1.5 \mu\text{m}$  transition. The spontaneous emission probability  $A_{JJ'}$  is proportional to the line strength  $S_{JJ'}$  using local field correction factor  $(n^2 + 2)^2/9$  through the relation

$$A_{JJ'} = \frac{64\pi^2 e^2}{3h(2J+1)\lambda_{\text{mean}}^3} n \left[ \frac{(n^2 + 2)^2}{9} \right] S_{JJ'}. \quad (3)$$

As shown in Fig. 2, both the  $S_{JJ'}$  and  $A_{JJ'}$  increased with the increase of  $\text{Al}_2\text{O}_3$  content up to 20 000 ppm and decreased over 20 000 ppm of  $\text{Al}_2\text{O}_3$ . The increase of  $S_{JJ'}$  and  $A_{JJ'}$  is considered to be caused by the increase of the electric-dipole component of  $S_{JJ'}$  because the magnetic-dipole component of  $S_{JJ'}$  is independent of the change of ligand field. On the other hand,  $\tau_f$  showed little  $\text{Al}_2\text{O}_3$  dependence and was about 10 ms, which is a typical value for the lifetime of the  $^4\text{I}_{13/2}$  level of  $\text{Er}^{3+}$ . Fig. 3 shows the nonradiative decay rate  $W_{\text{NR}}$  and quantum efficiency (QE). Using  $\tau_f$  and  $A_{JJ'}$ ,  $W_{\text{NR}}$  and QE are expressed as

$$W_{\text{NR}} = \tau_f^{-1} - \sum_{J'} A_{JJ'} \quad (4)$$

$$\text{QE} = \frac{\sum_{J'} A_{JJ'}}{W_{\text{NR}} + \sum_{J'} A_{JJ'}}. \quad (5)$$

### B. $\text{Al}_2\text{O}_3$ Dependence of $\Omega_6$ Parameters in Fibers

In order to investigate the  $\text{Al}_2\text{O}_3$ -content dependence of the  $\Omega_t$  ( $t=2,4,6$ ) parameters of doped  $\text{Er}^{3+}$  ions, we analyzed each absorption band that corresponded to the optical transition between  $4f$ -multiplets of  $\text{Er}^{3+}$  ions using the Judd–Ofelt theory.

Fig. 4 shows the comparison of the absorption spectrum of EDF ( $\text{Al}_2\text{O}_3$  content: 20 000 ppm) with that of an aluminosilicate bulk glass whose composition was the same as that of an aluminosilicate EDF sample ( $\text{Al}_2\text{O}_3$  content: 20 000 ppm).

We can see the four absorption bands corresponding to the transitions from the  $^4\text{I}_{15/2}$  to the  $^4\text{F}_{9/2}$ ,  $^4\text{S}_{3/2}$ ,  $^2\text{H}_{11/2}$ , and  $^4\text{F}_{7/2}$ . The absorption spectrum of the  $^2\text{H}_{11/2}$  is remarkably

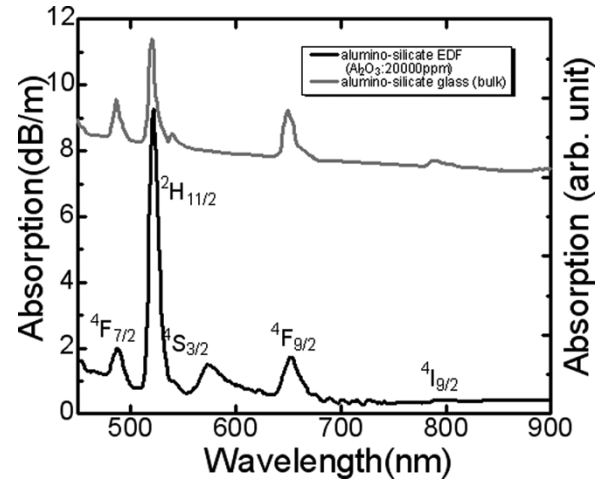


Fig. 4. Comparison of absorption spectra for  $\text{Al}_2\text{O}_3$ -doped EDF with an aluminosilicate bulk glass.

larger in aluminosilicate EDF than of those in aluminosilicate bulk glass. This enhancement of the absorption peak of the  $^2\text{H}_{11/2}$  was also observed in other erbium-doped fibers.

Fig. 5 shows the  $\text{Al}_2\text{O}_3$ -content dependence of the Judd–Ofelt intensity parameters  $\Omega_2$ ,  $\Omega_4$ , and  $\Omega_6$  calculated from the line strength absorption spectra in 400–1650 nm. The measured line strength could be matched with the best fit  $\Omega_t$  results to 10%–15% (rms) accuracy for all the aluminosilicate EDFs examined. As shown in Fig. 5, the  $\Omega_6$  parameter was found to increase with the increase of the  $\text{Al}_2\text{O}_3$  content up to 20 000 ppm. On the other hand, according to the Judd–Ofelt theory,  $S_{JJ'}$  was defined with the  $\Omega_t$  parameter as follows:

$$S^{\text{edf}}[S, L, J; S', L', J'] = \sum_{t=2,4,6} \Omega_t \times \left| \langle 4f^N S, L, J | U^{(t)} | 4f^N S', L', J' \rangle \right|^2 \quad (6)$$

where  $\langle 4f^N S, L, J | U^{(t)} | 4f^N S', L', J' \rangle$  denotes the reduced matrix element. The coefficients  $\Omega_2$ ,  $\Omega_4$ , and  $\Omega_6$  contain the effects of odd-symmetry crystal-field terms. In particular, the  $S_{JJ'}$  of the electric dipole components of the  $1.5\text{-}\mu\text{m}$  transition with  $\text{Al}_2\text{O}_3$  content is given by [6]

$$S^{\text{ed}} \left[ ^4\text{I}_{13/2}; ^4\text{I}_{15/2} \right] = 0.019 \Omega_2 + 0.118 \Omega_4 + 1.462 \Omega_6. \quad (7)$$

While  $S_{\text{ed}}$  is a function of glass structure and composition using the  $\Omega_t$  ( $t=2,4,6$ ) parameter,  $S_{\text{md}}$  is independent of the ligand fields and is characteristic to the transition determined by the quantum numbers. For the transition between the states which meet the transition selective rules  $\Delta S = \Delta L = 0$ ,  $\Delta J = 0$ , and  $\pm 1$ , there exists the contribution of magnetic dipole transition [7], [8]. Therefore, the increase of total line strength  $S_{JJ'}$  of the  $1.5\text{-}\mu\text{m}$  transition with the increase of  $\text{Al}_2\text{O}_3$  shown in Fig. 2 can be largely dominated and explained well by the increase of the  $\Omega_6$  intensity parameter shown in Fig. 5.

The value of the  $\Omega_6$  particularly tends to be affected by the basicity of glass and is known empirically to become larger as the basicity decreases [9]. Therefore, the increase of the  $\Omega_6$  parameter indicates that the increase of  $\text{Al}_2\text{O}_3$  weakens covalent

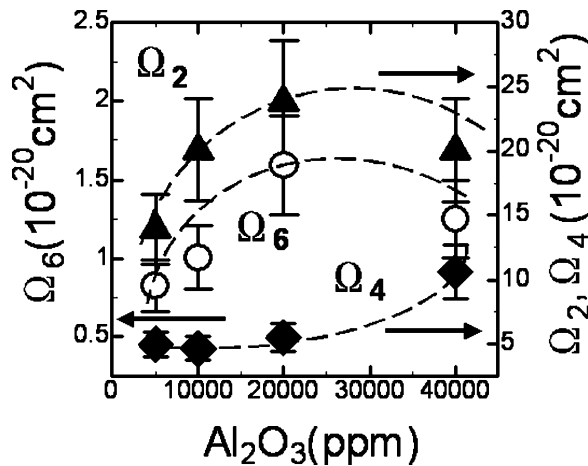


Fig. 5.  $\text{Al}_2\text{O}_3$ -content dependences of the Judd-Ofelt intensity parameter  $\Omega_2$ ,  $\Omega_4$ , and  $\Omega_6$ .

character of Er-O as a results the formation of Si-O-Al. Generally, it is known that the coordination number of  $\text{Al}^{3+}$  becomes smaller in the host glasses with higher basicity and the production of four-coordinated  $\text{Al}^{3+}$  is enhanced compared to the formation of six-coordinated  $\text{Al}^{3+}$  [10]. Moreover, the optical basicity of four-coordinated  $\text{Al}^{3+}$  in the network of  $\text{SiO}_4$  is known to be smaller than that of six-coordinated  $\text{Al}^{3+}$  (microscopic  $\lambda$  of Si-O-Al is 0.59 for four-coordinated  $\text{Al}^{3+}$  and 0.65 for six-coordinated  $\text{Al}^{3+}$ ) [11]. Therefore,  $\text{Er}^{3+}$  ions have the possibility to be surrounded by more four-coordinated  $\text{Al}^{3+}$  ions rather than six-coordinated  $\text{Al}^{3+}$  ions with an increase of  $\text{Al}_2\text{O}_3$ . The decrease of the basicity around  $\text{Er}^{3+}$  sites up to 20000 ppm of  $\text{Al}_2\text{O}_3$  content can be considered in particular as the result of the increase in the number of the four-coordinated  $\text{Al}^{3+}$  surrounds the  $\text{Er}^{3+}$  site.

#### C. $\text{Al}_2\text{O}_3$ Dependence of $\Omega_2$ Parameters in Fibers

As shown in Fig. 5, the value of the  $\Omega_2$  parameter in fibers takes about three times as large a value as those in aluminosilicate and borate bulk glasses [12] ( $\Omega_{2:(\text{Al}-\text{Si})\text{glass}} = 4.6$ ,  $\Omega_{2:(\text{Bo})\text{glass}} = 3.21$ ), which corresponds to the experimental results of a strong absorption band of the  ${}^2\text{H}_{11/2}$  as shown in Fig. 4. The relation between the  $\Omega_t$  ( $t = 2, 4, 6$ ) parameter,  $r$  and the electric-dipole line strength component of  ${}^2\text{H}_{11/2}$  is given by

$$S^{\text{ed}} \left[ {}^2\text{H}_{11/2}; {}^4\text{I}_{15/2} \right] = 0.7056 \Omega_2 + 0.4109 \Omega_4 + 0.0870 \Omega_6. \quad (8)$$

This relation shows that the electric-dipole line strength of the  ${}^2\text{H}_{11/2} - {}^4\text{I}_{15/2}$  is largely dominated by the value of the  $\Omega_2$  parameter. Therefore, the experimental results of a strong absorption band of the  ${}^2\text{H}_{11/2}$  can be explained well by (11). In case of  $\text{Er}^{3+}$ , the electric-dipole line strength of the  ${}^4\text{G}_{11/2} - {}^4\text{I}_{15/2}$  is also expected to show very strong absorption in fibers because the reduced matrix element  $\langle ||U^{(2)}|| \rangle$  of the  ${}^4\text{G}_{11/2} - {}^4\text{I}_{15/2}$  takes dominantly large value similar to that of the  ${}^2\text{H}_{11/2} - {}^4\text{I}_{15/2}$

$$S^{\text{ed}} \left[ {}^4\text{G}_{11/2}; {}^4\text{I}_{15/2} \right] = 0.9178 \Omega_2 + 0.5271 \Omega_4 + 0.1197 \Omega_6. \quad (9)$$

Both the  ${}^4\text{G}_{11/2} - {}^4\text{I}_{15/2}$  and the  ${}^2\text{H}_{11/2} - {}^4\text{I}_{15/2}$  are called the hypersensitive transition that is dominated and enhanced by the large  $\Omega_2$  parameter. It is known that the value of the  $\Omega_2$  is raised drastically by lowering the symmetry of the rare-earth ligand field [13]. According to the Judd-Ofelt theory, the  $\Omega_2$  critically dominates the hypersensitive transition that was reported to be largely affected by the polarized distortion of the ligand field [14], [15]. Therefore, the  $\text{Er}^{3+}$  sites in fibers are considered to have larger polarization or asymmetric distortion than in bulk glasses. Moreover, the  $\Omega_2$  also increased with the increase of the  $\text{Al}_2\text{O}_3$  content. Considering the  $\text{Al}_2\text{O}_3$  dependence of the  $\Omega_6$  parameter, this indicates that the substitution of six-coordinated  $\text{Al}^{3+}$  for four-coordinated  $\text{Al}^{3+}$  with the increase of  $\text{Al}_2\text{O}_3$  may occur. Therefore, high  $\text{Al}_2\text{O}_3$  doping is likely to enhance the distortion of the ligand field of  $\text{Er}^{3+}$  site caused by the existence of four- and six-coordinated  $\text{Al}^{3+}$ .

#### IV. CONCLUSION

The  $S_{JJ'}$  and the  $A_{JJ'}$  of the  $\text{Er}:1.5\text{-}\mu\text{m}$  transition increased with the increase of  $\text{Al}_2\text{O}_3$  content in EDF. The  $\Omega_6$  parameter increased with the increase of  $\text{Al}_2\text{O}_3$  content, which can be explained by the compositional dependence of the  $S_{JJ'}$  of the  $\text{Er}:1.5\text{-}\mu\text{m}$  transition. These results indicate that a decrease of local basicity occurs by adding  $\text{Al}_2\text{O}_3$ . The  $\Omega_2$  parameter in a fiber form was found to be about three times as large as those in a bulk glass form and also increased with an increase of  $\text{Al}_2\text{O}_3$  content. The decrease of the local basicity and the symmetry of the  $\text{Er}^{3+}$  ligand field in aluminosilicate EDF samples are attributed to the increase of four-coordinated  $\text{Al}^{3+}$ .

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